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Systems of order.

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SYSTEMS OF ORDER

CARNEGIE INSTITUTE
OF TECHNOLOGY
(M. S. DEGREE, 1950)

BY

P. H. McGANN

Thesis
M188

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[illegible] OF ORDER

[Carnegie Institute of Technology.]
[M.S. degree, 1950]

F. H. McDowell
1st December 1949

In an ordinary solid solution, the different species of atoms are arranged at random on the atomic positions of the lattices. At a given composition AB, any individual lattice point is occupied indifferently by either A or B atoms. Until comparatively recent times this purely random affair was thought to apply in all instances; no order in the atomic arrangement was surmised. However, in late years, it has been determined, largely through X-ray diffraction, that in a multitude of cases the atoms are arranged in as definite a way as are the two different elements of an ionic salt.

Strangely enough, the earliest prediction of ordered structure was not based on X-ray evidence, but on chemical experiments, by Tammann (322, 323). He found that suitably treated Cu-Au alloys of greater than 50 atomic % Au were resistant to HNO3, while less Au, the Cu atoms of the solution were dissolved but the Au atoms remained. On the basis of this evidence he concluded that a 50 atomic % Cu-Au alloy contained an ordered arrangement of atoms, and any extra Cu atoms did not fit in the arrangement so were readily removed.

Today, the final evidence of the existence of ordered arrangements, or superlattices, is furnished by the presence of "superstructure lines" on X-ray diffraction patterns. The earliest superlattice lines were observed by Bain (Ref. 23A of 239) for Cu_3Au and by Thomassen (Ref. 25B of 239) for $FeCo$. Johansson and Linde (Ref. 23A of 239) are responsible for the first analysis of an ordered structure from its X-ray pattern; they worked with the alloy $CuAu$.

Superstructures are characterized by the fact that the atoms of one crystal lattice are surrounded preferentially by atoms of the other type. It is well to bear in mind that, while in some cases the atoms surround themselves

as completely as is possible with unlike neighbors, in others they stop definitely short of this limit.

Superlattices form at relatively low temperatures. As the temperature increases, the first effect is only a greater amplitude of thermal vibration of the atoms about their equilibrium positions. When this effect becomes large enough, occasional pairs or small groups of atoms acquire sufficient energy to break away from their places in the lattice and interchange positions with each other. At all temperatures above a certain critical value the atomic randomness persists. As the temperature is lowered through the critical point order sets in, increasing as the temperature drops, and approaching perfection only at low temperatures.

Atoms go from an alloy by introducing into a pure metal atoms of a different metal. If the atoms of both types are then allowed to adjust their positions and crystal structure until stable, thermodynamical equilibrium is obtained. The free energy of the system reduces to its lowest value.

Atoms are said to attract one another if the internal energy of the crystal becomes lowered when the atoms are redistributed among the atomic sites in such a way as to increase the number of dissimilar atoms that form nearest neighbors. If dissimilar atoms attract each other to about the same extent as similar atoms, then each atom is relatively indifferent to the type of neighbors it possesses. The two kinds of atoms behave as if they were diluted and the mixture becomes homogeneous down to atomic proportions. A random solution is the result. If dissimilar atoms are attracted more than similar atoms, then the tendency is to form a structure in which atoms of one kind are surrounded by atoms of the other

kind, in order to obtain a maximum number of dissimilar atoms as nearest neighbors. In true metals, with similar electrochemical characteristics, the resulting structure is usually an ordered solid solution or superlattice.

Order can be subdivided into two categories. It can be described from a local point of view, with reference to the tendency of atoms to be surrounded by unlike neighbors. This phenomenon is referred to as short range order. If this localization extends for many interatomic distances in a lattice, atoms of one kind will be segregated on one set of atomic positions, leaving those of the other kind to the remaining positions. The result at worst can be described as a lattice of A atoms interpenetrating a lattice of B atoms. This segregation of atoms to particular sites takes place with little or no lattice deformation, but gives an ordered solid solution -- a superlattice displaying long range order.

It is evident that a perfectly ordered solution can only occur when the ratio of the numbers of different atoms in the alloy is a simple one, since the alternation of the dissimilar atoms through the structure demands an exact and simple ratio. Thus, today's well recognized superlattices are observed in either a 3:1 or 1:1 ratio. In systems showing superstructures at near ratios, partial or imperfect order is often obtained at compositions which differ slightly from the exact ratios. A superlattice can, of course, occur only when the alloy system forms a solid solution extending over a range of composition which includes at least one of these ratios, although fulfillment of this condition is no way assures formation of a superlattice at the critical compositions.

The formation of a superlattice may be regarded as a process in which the lattice strain produced by the solute element in the random solid solution is relieved by the formation of an ordered structure. Provided, then, that the solvent and solute atoms are of sufficiently similar size to permit the formation of a wide solid solution, the tendency to form superlattices increases with increasing difference in atomic diameters, since the greater this difference, the greater the strain to be relieved. Apart from this simple effect of atomic diameter, the formation of a superlattice is favored by increasing electrochemical factor. It is reasonable, therefore, that there is a continuous range, from superlattices involving only the metallic bond and formed merely to relieve the lattice strain resulting from atoms of unequal size, to superlattices with increasing degree of ionic bonding.

Theoretical treatments of superlattices have been made by a variety of investigators. The first work was the attempt of Bragg using formal thermodynamic relations as a basis. More recent work used simple assumptions about atomic forces and obtained quantitative results which compared with experiment quite favorably. Any critical analysis of this work would be quite extensive, so that mention of the most important theories only will be made.

Bragg and Williams used as the basic aspect of their theory, long range order. They assumed that V, the energy to effect an interchange of a B atom and an A atom, is proportional to the degree of long range order S, according to the equation $V = V_0 S$ where V_0 is a constant representing the interchange energy when long range order is absent. This assumption leads to a theory predicting a critical temperature above

times rich, sometimes without, a latent heat. The absence or short range order from this theory is a defect, since it seems certain that the principal interactions in crystals are between very close neighbors.

A logical development of superlattice theory is therefore to consider a concept of order that concerns only nearest neighbors, and to compute ordering energies based on nearest neighbor interactions. Theories based on this point of view have been advanced by Bethe and extended by several others, especially Fowler. As long distance order is defined in terms of the number of right atoms, so short range order is defined in terms of the number of right pairs (right pair being one of unlike atoms -- as AB pair).

With the extensive condensed theory of the order-disorder concept [illegible] it appeared [illegible] to compile a list of alloys which various authors have found to exhibit the ordering of a superlattice. The data presented is the result of a literature survey by this author covering from 1932 through the present date. Quite frequently, the paper from which the information was compiled gives but very sketchy data on the reaction. Such systems are generally mentioned only as a side issue in a paper on some subject quite remote from the principal topic of order-disorder reactions. Similarly, some systems listed have been reported as forming a superlattice by a single author, with no amplification by others. The scope of the present paper is all to extensive to be justify further investigation on alloys where complete data is lacking. Consequently, the author will merely present such material as is available without

comment as to its potential authenticity.

The systems will be compiled in alphabetical order, using the components with highest alphabetical sequence of presented English metallic name (not symbol), regardless of fractions present in the system.

Systems Ni-Al to Order

Cr-Al

A superstructure has been reported (Ref. 370 of 939) for tetragonal $CrAl_4$. This structure forms upon the body-centered cubic lattice. If we consider planes of the (100) type, which are formed alternately of cube corners and of cube centers, we find that every third plane contains Al atoms and the remaining planes Cr atoms.

Al-Co-Fe

A ternary phase structure was found (1E) in the Fe-Co-Al system with less than 60 atomic % Al somewhere from 0.60 %.

Ni-Co-Al

Equilibria have been shown (2A) in quaternary Ni-Co-Fe-Al alloys at Co ratios of 50:10, 50:30, 50:50, and 20:80. In this region there are two phases, β which the α' is FCC, ordered, and β is BCC, ordered. The α' phase in the Ni-Al system at about 13% Al extends to about 2% Co at 1000°C and 3.6 Co at 200° C in the binary system.

Al-Cu

A superstructure formed prior to the θ' phase in the Al-Cu. Cu-Al alloy is described (13E). This unit has a system of three tetragonal structures oriented with basal lines coincident with the (100) planes. It is suggested that an ordering of the Cu-Al planes

has occurred.

Cu-Mn-Al

The Heusler Alloy, Cu_2MnAl in the ferromagnetic condition is highly ordered (Ref. 3:3.3 F of 239). The ordered state results from a rapid cooling from a 600C anneal line temperature. The superlattice atoms are distributed in a BCC cell.

Fe-Al

As Al is added to the BCC Fe lattice, the Al atoms replace Fe atoms at random until a composition of 18% Al is reached. Here, the solute Al atoms cause a difficulness (Ref. 3A of 239) attributed to the onset of order in the annealed alloys. At 25% Al, slow cooling produces the ordered Fe_3Al lattice in which not only the nearest neighbor of an atom, but also its next nearest neighbors are unlike atoms. In the Fe_3Al lattice, Al atoms take such positions that, in following the BCC lattice along a cube diagonal, one meets Al atoms at alternate small cube (BCC) centers. On addition of further Al, the solute Al atoms begin to fill in the centers of the additional small cubes. At Fe Al, both rapidly cooled and annealed alloys acquire an ordered structure of the C. Cl type, with cube centers occupied by Al atoms and the corners by Fe atoms.

Fe-Ni-Al

There is also evidence of ordered and disordered phase constituents in 700 Alloys (.1). An alloy of Fe, Ni (10-30%), and Al (9-17%) quenched from 1100° - 1200° C, and annealed at 700C decomposes to $\lambda + \lambda'$, retained by an ordering of λ' (?).

is cooled from above 1100° C at a controlled rate decomposition of α into $\alpha + \alpha' + \gamma$ occurs at 7 00° C; below this temperature ordering of α' occurs.

Ni - Al

In an alloy of 31.5 atomic % Al, the Beta phase undergoes partial disordering at high temperatures and hence is fusible. The ordered structure is of the cubic Cs Cl type (1'9).

Al - Ag

The first stage (135) in precipitation in Ag-Al alloys is the collection of Ag atoms in nuclei with partial order in Al above.

Cu - Sb

Alloys with 60 - 64.0&% Cu consist of homogeneous Beta phase, a cubic structure with 16 atoms per unit cell; lattice parameter is 3.914 A. An orderly arrangement (3) resembling the Fe Al_3 structure is described. It corresponds to the formula $Cu_3 Sb$ ($Cu_3 Au$). Ordered arrangements of $Mn_3 Sb$ and $Mn_2 Sb$ have been described, but the author has no Sb-rich information on these structures.

Ni - Sb

The formation of a precipitate from the α phase is confirmed (288). The ordered structure α' ($Ni_{1.1}$ Sb) appears at 1000 C; its range of existence at room temperature is 88 to 90 atomic % Ni. The alloy is ferromagnetic.

Sb - Ag

A study of alloys, 17-31% Sb, is investigated (344). Two intermediate phases are confirmed, ε , a BCP lattice, and a rhombic

phase, roughly 4at% Be which exists at both high and low temperature modifications. The transition is completed at 410° - 430° C, depending on composition, and is interpreted as an order-disorder change.

Cu - Be

The disordered crystal is BCC; the ordered structure is a cubic of the Cs Cl type.

Au - Cd

As above, the disordered crystal is BCC; the ordered crystal is of the cubic Cs Cl type.

Mg - Cd

Below 260° C, superstructures appear at the compositions Cd_3 Mg, Cd Mg, and Cd Mg_3 (2). Superlattices have been found with Cd ranges from 70.12% to 93.1% (167). Both the Mg_3 Cd and Mg Cd_3 ordered structures possess the CPH lattice.

Mg - Ag - Cd

The quasibinary system, Mg-Ag Cd_3, exhibits a CPH ordered structure in the range 37-70 atomic % Mg.

Mn - Cd - Sn

A superlattice has been reported (167) at the composition 72% Cd and 10% Mn.

Si - Cr

Electrical and dilatometric measurements in the Si-rich Cr-Si solid solution exhibit an anomaly concluded to be due to an order-disorder transformation similar to that in the Cu-Sn -Mn solid solution (305).

Cr - Pt

A superstructure of unit is thought to be Cr_3Pt has been reported (107). Another author (110) finds A superstructure in alloys of 37-41.6 atomic % Cr on slow cooling from 1330° C.

Fe - Co

These alloys, in the composition range 40-65 % Co, order from a BCC to a cubic of the Cs Cl type. T_c = 730° C (91). Ordered alloys exhibit a larger lattice constant at room temperature than quenched, disordered, material, owing to the lower symmetry of the ordered alloy (96).

Co - Pt

One investigator (174) finds that a cast sample of Co Pt contains a mixed structure, FCC (a = 3.75), and BCC (a = 2.85) similar to the Cs Cl cell. A quench from 1200° C results in FCC structure alone. A volume increase of 11% when BCC forms from FCC is the author's basis for concluding that the BCC must become ordered. More recent papers are probably more accurate. There is general agreement that the disordered lattice is a FCC with a_o = 3.751 - 3.744, and the ordered structure is an Au Cu -- like FCT lattice with a_o = 3.785 - 3.818 and c = 3.639 - 3.608 (109,163). T_c = 825°C. This work relates to a 50 - 50 atomic % composition.

Au - Cu

It is on this best known of all ordering systems that most work has been published. Disordered alloys of this system possess a FCC structure. In the Cu Au range, a superlattice forms in which alternate layers of Cu and Au atoms form on (001) planes on the FCC

solid solution lattice, disordering it into a tetragonal structure. T_c for this reaction is about 375° C (117). Between 370° and 408° C, another structure, referred to as Cu Au II, is stable. It is an orthorhombic lattice, with two nearly equal axes, and the third about ten times as long (182). At Au Cu is found another ordering process. FCC disordered to FCC ordered (250). Ordered Au Cu and Au Cu$_3$ are related by an intermediate series of phases (289). At 75 atomic % Cu there exist indications of long range order at 1960 C (116). At 370° C. 0.7% of the Au Cu lattice, 0.1% of the Au Cu$_3$, and 4% of the $AgAu$ Cu$_3$ are not in ordered positions (345). The critical points of these alloys are raised by pressure (340).

Au-Cu-Bi

High concentrations of Bi do not affect transformations of the Au-Cu system. The effect of Bi on Au Cu$_3$ ordering is less than with the Au Cu lattice (211).

Au-Cu-Ag

Addition of Ag lowers the critical temperature of the Au-Cu system (169,184). Addition of 50 at. % Ag inhibits ordering completely (51).

Cu-Mn-I

The ternary phase $AgAu$ Mn I shows a complex order-disorder change resembling somewhat to t in Au Cu, but involving ordered and disordered arrangements of both metal atoms and normal lattice points.

Cu-Si-Sn

Recrystallization in a 10% Cu 20% Si, 20% Sn alloy is considered to be due to an ordering process, from a cubic to a FCC lattice (279)

Cu-Mn-Sn

With greater than 20 atomic % Mn, the Beta phase has an ordered structure which, for a Cu$_2$ Mn Sn alloy, persists at 630° C (54).

Cu-Pd

Two structures in this system have been found to exhibit ordering. Cu Pd and Cu$_3$ Pt. Cu Pd orders to a cubic CsCl type from the disordered BCC lattice, Cu$_3$ Pt in the range 37-48 atomic % Pd, in which the CsCl cubic forms from a disordered FCC. Cu$_3$ Pd orders from an FCC to an ordered FCC. The latter is slightly distorted from the cubic, investing it with tetragonal symmetry (45).

Cu-Pt

In this system near the 50 atomic % composition, the FCC disordered lattice becomes ordered rhombohedral with alternate planes of Cu and of Pt atoms on (111) planes. This structure has trigonal symmetry (Ref. 27 on 239,248). A AA stack of Pt (298) and Cu$_3$ Pt$_2$ have been found to agree with this concept. Order in the composition range 5-95 Atomic % Pt exists only below 615°C, with the maximum critical temperature at 20 atomic % Pt (93). The lowest degree of disorder exists in alloys containing a slight excess of Pt over the Cu$_3$ Pt composition. Alloys between 37 and 48 atomic % Pt change from a disordered FCC to the Beta Cubic type structure, ordered BCC. Pt Cu$_3$ changes from a disordered to ordered FCC lattice. One author writes (93) that ordered alloys exist between 30 and 90 atomic % Pt, while another (265) believes no order exists above a composition of 60 Pt. Degrees of disorder (93) have been as follows: Cu Pt, Cu$_3$Pt, Cu Pt$_3$.

Cu-Ga

In alpha brass, short range order is persistent up to the melting point (144). The alpha prime lattice is ordered, being similar to the 50 at % Cu alloy (FCC to ordered simple tetragonal). At room temperature, alpha prime exists a long time, changing to a stable alpha on tempering (184). Disordered beta brass is a random BCC lattice. On ordering, a Cs Cl type BCC is produced in which a_o = 2.9 ± Å (1). T_c is about 460° C.

Au-Al

This system forms a BCC of the Cs Cl type on ordering from a disordered BCC.

Au-Pb

The alloy $Au Pb_2$ is highly ordered near the fusion temperature (199).

Au-Tl

This is another system exhibiting a high degree of order near the melting point (199).

Au-Zn

The beta phase, with limits of homogeneity of 47.5 to 53.0 at % Au, has Cs Cl type structure of 2 atoms per unit cell, with maximum order at equi-atomic percentages of Zn and Au (1). The order-disorder transformation in an heating above 500° C, but does not proceed to completion up to the melting point of 725° C.

Ag-Mg-Al

The $Ag_3 Mg Al$ Heusler phase shows a complex order-disorder behavior resembling ordering in Cu$_3$-

Fe-Ni

Experimental evidence indicates a superstructure in the neighborhood of Ni_3Fe (193). Superlattice lines at 74.3 atomic % Ni have been found (288). T_c is around 5000 C, but the transformation is remarkably sluggish. The reaction is probably of the discontinuous type (181). The disordered lattice is FCC, as is the ordered product.

Mn-Fe-Ni

The alloy $(Mn_3Fe)Ni_3$ exhibits an ordering reaction in changing from a random FCC to an ordered FCC.

Fe - Pd

On annealing below 650° C, $FePd$ forms an ordered tetragonal with an axial ratio of less than unity. The superlattice is most stable with an excess of Pd. At 60 atomic % Pd, it is stable up to 775° C, while at 50 it is only to about 675° C at 51.9% Pd. There exists also an $FePd_3$ ordered state with the same structure (FCC) as $AuCu_3$ (164).

Fe-Pt

The disordered FePt exists as a FCC and at about 1300° C (314), orders to the AuCu type tetragonal face centered structure.

Fe-Pt-Rh

An alloy of 50 atomic % Fe, 45 atomic % Pt, and 5 atomic % Rh undergoes the tetragonal structure of the CuAu type (Ref. 360 or 239).

Fe-Rh

At 5 atomic % Rh, this alloy forms an ordered structure (235).

Fe-Si

Some magnetic measurements (Ref. 328 or 239) indicate ordered phases at 12.5 and 21 atomic % Si. X-ray evidence (Ref. 328 or 239) establishes only the latter, the Fe_3Si ordered phase, which is a BCC of the Cs Cl type.

Au-Mn

The beta phase occurs throughout the whole range (0-100) but ordering is most complete in the region of 50 atomic % Mn. There is some evidence, but no full investigation, for a superlattice in the alpha phase in the 21 atomic % Mn region. The beta phase ordered from a BCC to a BCC of the Cs Cl type.

Ni-Mn

The composition $MnNi_3$ orders from a random FCC to an ordered FCC. The transition temperature has not been agreed upon by investigators. Estimates include 410° C (326), 420° C (327), less than 950° C (334), and 300-400°C (185) with the remark that the transition occurs over a range, so real critical temperature existing. One author reports the formation of a FCT with axis at 1050° C (130).

Pd-Mn

A superlattice, Pd_3Mn at 10% Mn has been reported (130). The transition sequence observed were: FCC --> 1165° C --> tetragonal --> <300° C --> ordered tetragonal.

Au-Mn

Superstructures have been reported for compositions of Au_3Mn, $AuMn_3$ and $AuMn_4$. No qualifying data was found.

Ni-Pt

A stoichiometric, ordered Ni Pt phase was observed (92), with a 1000 C limits of 50-55 atomic % Pt.

Ni-Sn

At 900° C occurs an order-disorder transformation in the beta phase. Ni3 Sn is the superstructure; it is a CCP of the Mg Ni type.

Au-Pt

In the region 20-30 atomic % Pt there is a new phase, Au3 Pt, which at less than 600° C has a simple ordered structure; at greater than 600° C it becomes disordered (252).

Au-Sn

In the range 28-34 % Sn is a change of disordered to ordered lattice (Beta Beta one) taking place at 200-225° C. Similar to the Beta brass structure, this alloy exhibit the ordered BCC of the Cs Cl type.

As a final project, let us look briefly to the effect of the order-disorder phenomena on various properties of an alloy.

Since the ordered state is the one of lowest energy, energy must be supplied to create disorder. During disordering, the speed is heat is greater than normal; the value at one temperature depends on the perfect degree of order with temperature. A latent heat is displayed by some systems, not by others. Theory is weak in predicting this phenomenon.

Electrical resistivity is affected by ordering. If the disordered state is obtained at a temperature below the critical by a quench, the scoring from the distortion in resistivities are superimposed upon the discontinuous thermal behavior, and since/below

to electrical resistance. Annealing induces order, lowering the resistivity.

Magnetic properties are order-dependent. The Heusler alloys become ferromagnetic in the ordered state. Diamagnetic susceptibility increases for some alloys (e.g., Au CuZ, Cu_3Pd) upon ordering while it does not in others (e.g., Au_3Cd).

Whether of the long or short-range type, ordering is always accompanied by a dilatation or volume.

Young's modulus increases with ordering in Cu_3Pd and $AuCuZ$, but decreases in Au Cu and $CuPd$. Elastic anisotropy also is sensitive also to local order.

Mechanical properties are also altered when ordering occurs. Hardness, tensile strength, and the elastic limit are usually increased.

Plastic deformation tends to destroy long-range order.

Any paper attempting a complete analysis of the subject, order-disorder, would of necessity fill volumes. Our aim is to see that knowledge and theory of this phenomenon is at least in its adolescence stage. Modern work is, however, making rapid strides toward completion of theory and development, and given courage of completion it is to years a workable knowledge of the crystalline ordered.

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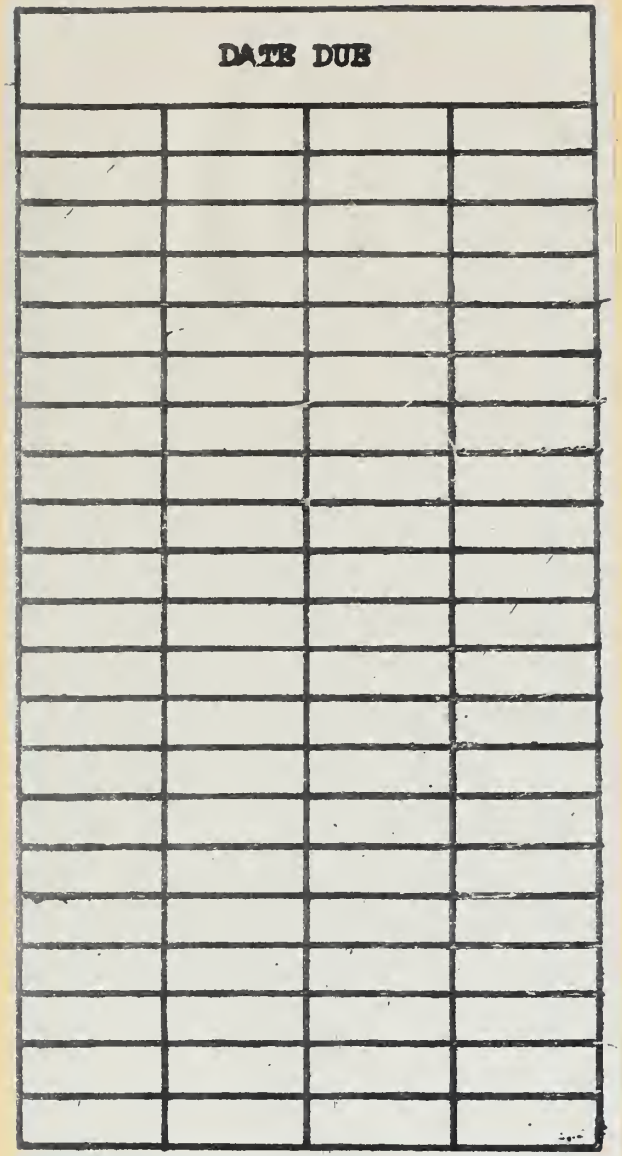
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